Reduction of Pauli paramagnetic pair-breaking effect in antiferromagnetic superconductors

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Antiferromagnetic superconductors in a magnetic field are studied. We examine a mechanism which significantly reduces the Pauli paramagnetic pair-breaking effect. The mechanism is realized even in the presence of the orbital pair-breaking effect. We illustrate it using a three-dimensional model with an intercalated magnetic subsystem. The upper critical field is calculated for various parameters. It is shown that the upper critical field can reach several times the pure Pauli paramagnetic limit. The possible relevance to the large upper critical field observed in the heavy fermion antiferromagnetic superconductor $CePt_3Si$ discovered recently is briefly discussed. We try to understand the large upper critical field in the compound $CePt_3Si$ and field-induced superconductivity in the compound $CePb_3$ within a unified framework.

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In this paper, we examine superconductivity in the presence of a coexisting antiferromagnetic long-range order. In a magnetic field, some types of antiferromagnet exhibit canted spin structures, which give rise to a net ferromagnetic moment proportional to the magnitude of the magnetic field. The ferromagnetic moment creates an exchange field on the conduction electrons by the Kondo interaction. When the Kondo interaction is antiferromagnetic, the exchange field reduces the effective Zeeman energy. As a result, the Pauli paramagnetic pair-breaking effect [1, 2] can be significantly reduced [3, 4], and the upper critical field H_{c2} can largely exceed the Pauli paramagnetic limit H_P (Chandrasekhar and Clogston limit).

The analogous compensation mechanism in superconductors with uniformly aligned spins is known as the Jaccarino-Peter mechanism [5], which explains the field-induced superconductivity (FISC) in the compounds $Eu_xSn_{1-x}Mo_6S_8$ [6], $CePb_3$ [7], and λ-$(BETS)_2FeCl_4$ [8, 9, 10, 11]. However, the resultant phase diagrams of the present mechanism are quite different from that of the Jaccarino-Peter mechanism. The superconductivity occupies a large single area including the zero field ($H = 0$) in the T-H phase diagram, while depending on the parameter values, the FISC may occur, where T and H denote the temperature and the magnetic field, respectively. In our previous papers [3, 4], we have proposed this mechanism in a multilayer system in a parallel magnetic field. We have adopted such a system, because the reduction mechanism is clearly illustrated. In this paper, we extend the same mechanism to more general systems in the presence of the orbital pair-breaking effect. The magnitude and the temperature dependence of the resultant upper critical field can be similar to those of the spin-triplet superconductivity of equal spin pairing.

The present work was motivated by the heavy fermion antiferromagnetic superconductor $CePt_3Si$ discovered recently [12]. Due to the heavy quasi-particle mass, the orbital pair-breaking effect is much weaker than in conventional metals. Furthermore, the compound exhibits antiferromagnetic transition at $T_{AF} \approx 2.2\,\mathrm{K}$ and superconducting transition at $T_c^{(0)} \approx 0.75\,\mathrm{K}$ at the zero field. The upper critical field $H_{c2}(0) \approx 5\,\mathrm{T}$ at $T = 0$ is much larger than the Pauli paramagnetic limit estimated by the simplified formula $H_P \approx 1.86[\mathrm{T/K}] \times T_c^{(0)}[\mathrm{K}] \approx 1.4\,\mathrm{T}$. Such a large H_{c2} seems to suggest spin-triplet pairing, although the lack of inversion symmetry of this compound seems disadvantageous to spin-triplet pairing. Recently, Frigeri *et al.* have shown that spin-triplet pairing is not entirely excluded by the lack of inversion symmetry. Therefore, equal spin pairing may be the reason for a large H_{c2} [13]. In this paper, however, we propose another scenario based on antiparallel spin pairing including spin-singlet pairing. Samokhin *et al.* have carried out band structure calculations and proved within their theory that the order parameter must be an odd function of the momentum $\boldsymbol{k}$ [14]. We should note that this does not mean the occurrence of equal spin pairing [15]. Hence, our scenario does not contradict their result.

In principle, we might be able to test these scenarios by rotating the magnetic field. The upper critical field H_{c2} must be highly anisotropic, if the large H_{c2} is due to equal spin pairing and the $\boldsymbol{d}$-vector is fixed. For example, in the compound $CePt_3Si$, the strong spin-orbit interaction favors a particular $\boldsymbol{d}$-vector [13]. For the magnetic field parallel to the $\boldsymbol{d}$-vector, the upper critical field must be strongly suppressed. In contrast, the upper critical field can be nearly isotropic in the present mechanism. The Knight shift measurement might also give useful informations [15].

As mentioned above, the purpose of this study is to examine the present mechanism in the presence of the orbital pair-breaking effect. Therefore, we examine a three-dimensional system with an intercalated magnetic subsystem. We divide the magnetic subsystem into two sublattices, which we call A and B. We approximate the conduction electron system with an isotropic continuum system for simplicity.

When we apply the theory to the compound $CePt_3Si$,

the A and B sublattices are alternate layers of Ce atoms, and the spins on each layer are ferromagnetically ordered [16]. The superconductivity and the antiferromagnetism may exist in the same degrees of freedom in this compound, but we simplify the situation by dividing the degrees of freedom into two coupled systems. We also ignore the spin-orbit interaction and the lack of inversion symmetry for simplicity, because they do not affect the present mechanism.

First, let us describe the spin structure in the magnetic subsystem. We consider the situation in which the antiferromagnetic transition occurs at a temperature much higher than the superconducting transition temperature, which is consistent with the observation in the compound $CePt_3Si$ [12]. Since the ordered state is rigid, we neglect the modification of the magnetic structure by the occurrence of superconductivity. Therefore, we consider the cases in which the magnetic subsystem is effectively described by the Hamiltonian

$$H_s = \sum_{i,j} J_{ij}\, \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \sum_i g_s \mu_B \boldsymbol{H} \cdot \boldsymbol{S}_i, \tag{1}$$

where g_s and μ_B denote the g-factor of the localized spins and the Bohr magneton [17]. We define z as the number of antiferromagnetic bonds for a given site. We set $J_{ij} = 0$ except for the nearest neighbor sites (i, j). For the compound $CePt_3Si$, we assume that $J_{ij} = J_{\parallel} < 0$ for (i, j) on the same layer (the same sublattice), while $J_{ij} = J_{\perp} > 0$ for (i, j) on the adjacent layers (the different sublattices). In this case, we have $z = 2$. We could examine other crystal structures. For example, we have $z = 6$ in the system on the cubic lattice. In any case, we assume that $J_{ij} = J > 0$ for the antiferromagnetic bonds.

We take the x-axis of the spin space as being in the direction antiparallel to the external field. Hence, we set $\boldsymbol{H} = (-H, 0, 0)$. The directions of the axes of the spin space do not necessarily coincide with the crystal axes in the present model. For $T \lesssim T_c^{(0)} \ll T_{AF}$, we neglect the thermal fluctuations and regard the localized spins as classical variables. If we ignore the anisotropy energy of the localized spins in the magnetic layers, the sublattice magnetization appears in the direction perpendicular to the applied field, since it is energetically favored. Hence, we introduce the classical variable θ by

$$\boldsymbol{S}_i = \bar{S}\,(\sin\theta, 0, \pm\cos\theta), \tag{2}$$

where we define the z-axis of the spin space as being in the direction of the sublattice magnetization. For the double sign $\pm$ in eq. (2), we adopt $+$ and $-$ signs when site i belongs to the A and B sublattices, respectively. Here, we have introduced the magnitude of the localized spins $\bar{S}$ taking into account the shrinkage of spins by fluctuations. The total energy of the magnetic subsystem is expressed as

$$E(\theta) = N\bar{S}\left[-zJ\bar{S}\cos 2\theta - 2h_s \sin\theta\right] + E_0, \tag{3}$$

where E_0 is a constant and $h_s \equiv g_s\mu_B H/2$. The total energy becomes minimum when

$$zJ\bar{S}\sin\theta = h_s \tag{4}$$

for $h_s/zJ\bar{S} \le 1$, *i.e.*, $H \le 2zJ\bar{S}/g_s\mu_B \equiv H_{AF}$. For a high field in which $H \ge H_{AF}$, the staggered moment disappears ($\theta = \pi/2$). From now on, we mainly consider a weak field $H \le H_{AF}$, where the antiferromagnetic order persists. The localized spins are also described by the spin density

$$\boldsymbol{S}(\boldsymbol{r}) = \sum_i \delta^{(3)}(\boldsymbol{r} - \boldsymbol{R}_i)\, \boldsymbol{S}_i, \tag{5}$$

where $\boldsymbol{R}_i$ denotes the position of site i.

We treat the degrees of freedom of the conduction electrons as a continuum model. The net spin moment $\bar{S}\sin\theta$ gives rise to the exchange field on the conduction electrons. In order to reproduce the exchange field, we employ an extended Kondo Hamiltonian defined as

$$\begin{aligned} H_K = \int d^3\boldsymbol{r} \int d^3\boldsymbol{r}'\, \hat{J}_K(\boldsymbol{r} - \boldsymbol{r}') \\ \times \boldsymbol{S}(\boldsymbol{r}) \cdot \sum_{\sigma\sigma'} \left[\psi_\sigma^\dagger(\boldsymbol{r}')\boldsymbol{\sigma}_{\sigma\sigma'}\psi_{\sigma'}(\boldsymbol{r}')\right], \end{aligned} \tag{6}$$

where $\boldsymbol{\sigma}_{\sigma\sigma'}$ and $\psi_\sigma(\boldsymbol{r})$ denote the Pauli matrices and the annihilation operator of the conduction electron at $\boldsymbol{r}$. Here, we note that eq. (6) is used in the calculation of the order parameter $\Delta(\boldsymbol{r})$, which appreciably varies in the length scale of the coherence length $\xi \gg a$, where a denotes the lattice constant. Therefore, after inserting eq. (5) in eq. (6), we replace the spin variable $\boldsymbol{S}_i$ by the spatial average $\langle \boldsymbol{S} \rangle_{\xi i}$ over the volume $v_{\xi i}$ on the order of ξ^3 near site i, which is defined as $\langle \boldsymbol{S} \rangle_{\xi i} \equiv \sum_{\boldsymbol{R}_j \in v_{\xi i}} \boldsymbol{S}_j \,/\, \sum_{\boldsymbol{R}_j \in v_{\xi i}} 1$. Thus, we obtain

$$\begin{aligned} H_K \approx \int d^3\boldsymbol{r}' \sum_i \hat{J}_K(\boldsymbol{R}_i - \boldsymbol{r}') \\ \times \langle \boldsymbol{S} \rangle_{\xi i} \cdot \sum_{\sigma\sigma'} \left[\psi_\sigma^\dagger(\boldsymbol{r}')\boldsymbol{\sigma}_{\sigma\sigma'}\psi_{\sigma'}(\boldsymbol{r}')\right]. \end{aligned} \tag{7}$$

In actuality, the microscopic fluctuations omitted here may affect the superconductivity through the self-energy of the electrons. We regard those corrections to the normal state as being included in the effective mass m^*, if it exists. Since $\xi \gg a$, there are many localized spins in the volume $v_{\xi i}$, the antiferromagnetic components of the spins vanish on average, while the ferromagnetic moment remains. Therefore, we obtain $\langle \boldsymbol{S} \rangle_{\xi i} \approx (\bar{S}\sin\theta, 0, 0)$. For specific lattice structures, this equation is satisfied exactly. Furthermore, we assume that the localized spins on z_K lattice sites around $\boldsymbol{r}'$ take part in the Kondo interactions with the conduction electron at $\boldsymbol{r}'$, and that all of these coupling constants are equal to J_K. Therefore, we obtain

$$H_K \approx h_{ex} \int d^3\boldsymbol{r}' \sum_{\sigma\sigma'} \left[\psi_\sigma^\dagger(\boldsymbol{r}')\sigma^x_{\sigma\sigma'}\psi_{\sigma'}(\boldsymbol{r}')\right], \tag{8}$$

where h_{ex} denotes the exchange field $h_{ex} = z_K J_K \bar{S} \sin\theta$. From eq. (4), we obtain

$$h_{ex} = \frac{g_s \mu_B}{2} \frac{z_K J_K}{zJ} H, \tag{9}$$

for $H \leq H_{AF}$. For $H \geq H_{AF}$, we have $h_{ex} = z_K J_K \bar{S}$.

Taking into account the exchange field h_{ex} and the external field h, the kinetic energy term of the conduction electron Hamiltonian is written as

$$H_0 = \sum_{\sigma\sigma'} \int d^3\boldsymbol{r}\ \psi^\dagger_\sigma(\boldsymbol{r}) \Big[\frac{-\hbar^2}{2m^*} \big(\boldsymbol{\nabla} - \frac{e}{c}\boldsymbol{A}(\boldsymbol{r})\big)^2 \delta_{\sigma\sigma'} - (1-\delta_{\sigma\sigma'})\,\tilde{h} \Big] \psi_{\sigma'}(\boldsymbol{r}), \tag{10}$$

where m^* and $\tilde{h}$ denote the band effective mass and the renormalized Zeeman energy $\tilde{h} = h - h_{ex}$, respectively. Here, we have defined $h = \mu_e H$ and $\mu_e = g_e \mu_B/2$ where g_e denotes the g-factor of the conduction electrons. For $H \leq H_{AF}$, we have

$$\tilde{h} = \big(1 - \frac{g_s z_K J_K}{g_e z J}\big)\, h = \big(1 - \frac{J_K}{J'_{AF}}\big)\, h \tag{11}$$

obtained by eq. (9), where we have defined $J'_{AF} \equiv g_e z J / g_s z_K$. In contrast, for $H \geq H_{AF}$, we have $\tilde{h} = h - z_K J_K \bar{S}$

In eq. (11), it is explicit that the Zeeman energy is reduced by the exchange field when $J_K > 0$. It is found that the reduction effect becomes maximum when $J_K = J'_{AF}$. Obviously, this reduction mechanism is not affected by the anisotropy of the order parameter. Hence, we consider the s-wave superconductor as an example.

Applying the standard theory of superconductivity to the present system, we obtain the integral equation for the superconducting transition temperature T_c [18, 19, 20, 21, 22]

$$\ln\frac{T_c}{T_c^{(0)}} = \frac{1}{2}\int_0^\infty dx \frac{1}{\sinh x} \int_0^\pi d\theta \sin\theta \times \Big[\exp\big(-\frac{\kappa'}{4}x^2\sin^2\theta\big)\cos(2\tilde{h}'x) - 1\Big], \tag{12}$$

where $T_c^{(0)}$ denotes the zero-field transition temperature, and we have defined $\kappa' \equiv (v_F/2\pi T_c)^2 2|e|H/c$ and $\tilde{h}' \equiv \tilde{h}/2\pi T_c$. Here, we have ignored the possibility of the Fulde-Ferrell-Larkin-Ovchinnikov state. The extension to include it is straightforward [4, 22]. From the form of κ', it is convenient to define a constant

$$a_m \equiv \big(\frac{v_F}{2\pi T_c^{(0)}}\big)^2 \frac{2|e|}{c}, \tag{13}$$

with which we can write $\kappa' = a_m H (T_c^{(0)}/T_c)^2$. We also define the ratio of the scales of the paramagnetic and orbital effects as

$$r_m \equiv \frac{h/2\pi T_c^{(0)}}{a_m H} = \frac{g_e \pi T_c^{(0)}}{8\epsilon_F}\frac{m^*}{m}, \tag{14}$$

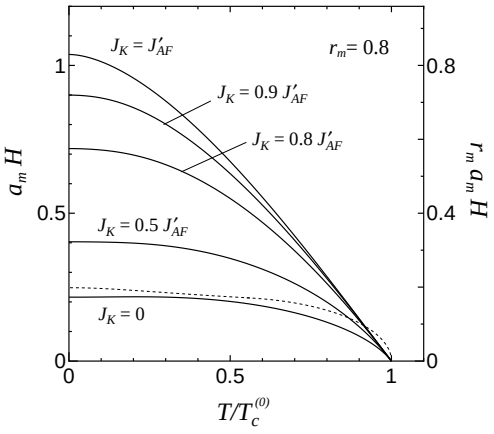

FIG. 1: Temperature dependence of upper critical fields for $r_m = 0.8$. The dotted curve shows the pure Pauli paramagnetic limit [23] in the absence of the orbital pair-breaking effect at $J_K = 0$, where $g_e = 2$. The curves of the upper critical field for $J_K/J'_{AF} = 1.1$, 1.2, and 1.5 coincide with those for $J_K/J'_{AF} = 0.9$, 0.8, and 0.5, respectively.

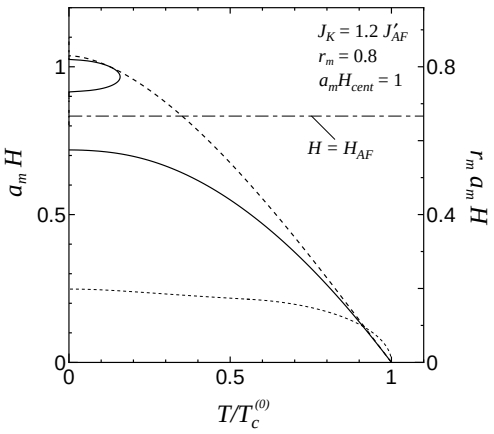

FIG. 2: Critical fields of superconductivity for $J_K = 1.2 J'_{AF}$, $r_m = 0.8$ and $a_m H_{cent} = 1$. The dotted and short dashed curves show the pure paramagnetic and pure orbital limits, respectively. The dot-dashed line shows the critical field of the antiferromagnetic phase.

where $\epsilon_F \equiv m^* v_F^2/2$.

Equation (12) is solved numerically. Figure 1 shows the results for $r_m = 0.8$ and $J_K < J'_{AF}$. It is found that the upper critical field is enhanced as J_K increases up to J'_{AF}. When $J_K \neq 0$, the upper critical field H_{c2} can exceed the Pauli paramagnetic limit $H_P(T)$. In particular, for $J_K \sim J'_{AF}$, the upper critical field H_{c2} can reach fourfold the Pauli limit H_P. When $J_K = J'_{AF}$, the pure orbital limit $H_{c20}(T)$ is recovered. For larger r_m, *i.e.*, weaker

orbital effect, the ratio H_{c2}/H_P increases.

If there is a temperature region in which $H_{c2}(T) > H_{AF}$ in Fig. 1, the curve of $H_{c2}(T)$ should be modified there so that it saturates more rapidly, because the exchange field is constant for $H > H_{AF}$. In the scale of the right vertical axis, $H = H_{AF}$ gives $r_m a_m H_{AF} = g_e z J\bar{S}/2\pi g_s T_c^{(0)}$. Hence, if $g_e/g_s \gg 1$ or if $zJ\bar{S}/T_c^{(0)} \sim T_{AF}/T_c^{(0)} \gg 1$, the upper critical field $H_{c2}(T)$ does not exceed H_{AF} in the whole temperature region.

In a high-field region in which $H \geq H_{AF}$, where the spins are aligned uniformly, the mechanism is reduced to the Jaccarino-Peter mechanism. In this case, the Zeeman energy completely vanishes at $H = z_K J_K \bar{S}/\mu_e \equiv H_{cent}$. Therefore, if $H_{cent} + H_P > H_{AF}$ is satisfied, we need to examine the possibility of FISC. Since $H_{cent}/H_{AF} = J_K/J'_{AF}$, FISC occurs only when $J_K \gtrsim J'_{AF}$ [3]. Figure 2 shows the critical fields for $J_K = 1.2J'_{AF}$, $r_m = 0.8$ and $a_m H_{cent} = 1$. This set of the parameter values gives $a_m H_{AF} \approx 0.83$, and if $g_s = g_e = 2$, $T_{AF} \sim zJ\bar{S} = 2\pi g_s T_c^{(0)} r_m a_m H_{AF}/g_e \sim 4\,T_c^{(0)}$. In Fig. 2, we find an area of FISC inside the region of $H_{c20}(T) > H > H_{AF}$.

In this context, the compound $CePb_3$ corresponds to the case of $H_{cent} \gg H_{AF}$, *i.e.*, $J_K \gg J'_{AF}$, and $r_m \gg 1$ [24]. In contrast, the compound $CePt_3Si$ has two possibilities, *i.e.*, $J_K \gtrsim J'_{AF}$ and $J_K \lesssim J'_{AF}$. For the former case, FISC might be observed at very high fields, if $H_{c20}(0) \gtrsim H_{cent} \gtrsim H_{AF}$ [25]. In the compound UPd_2Al_3, it was observed that $T_{AF} = 14.3\,\mathrm{K}$ and $T_c^{(0)} = 2.0\,\mathrm{K}$ [26]. Such a high T_{AF} suggests small J_K/J'_{AF}, so that the reduction of the paramagnetic effect is not pronounced. This coincides with the experimental fact that $H_{c2} < H_P$ and the absence of FISC in the compound UPd_2Al_3. Generally speaking, the difference in the experimental results $T_{AF} \approx 1.1\,\mathrm{K}$, $2.2\,\mathrm{K}$, and $14.3\,\mathrm{K}$ in $CePb_3$, $CePt_3Si$, and UPd_2Al_3, respectively, is consistent in the present theory with their phase diagrams, if their J_K's are of the same order.

Here, the condition $r_m \sim 1$ can be satisfied in heavy-fermion superconductors, but not in conventional metals, because $r_m \propto g_e T_c^{(0)}/m{v_F}^2 = g_e(T_c^{(0)}/\epsilon_F)(m^*/m)$. Also in the scenario with equal spin pairing, we need r_m of order 1, in order to reproduce a large ratio of H_{c2}/H_P as observed.

In conclusion, we have shown that the Pauli paramagnetic pair-breaking effect can be considerably reduced in antiferromagnetic superconductors, even in the presence of the orbital pair-breaking effect, unless $r_m \ll 1$. One of the necessary conditions for the occurrence of the present mechansim is that the Kondo exchange coupling constant J_K is positive and comparable to the scale of the antiferromagnetic exchange energy J'_{AF}. The present result may explain the high H_{c2} observed in the compound $CePt_3Si$. The phase diagrams of the compounds $CePt_3Si$ and $CePb_3$ can be understood within a unified framework. The positive J_K can be attributed to superexchange or kinetic exchange processes. Hence, the condition $J_K \sim J \sim J'_{AF}$ can be satisfied, because the antiferromagnetic exchange interaction J originates from exchange processes similar to those for J_K in the same crystal structure. The hybrid ruthenate-cuprate compound $RuSr_2GdCu_2O_8$ [27, 28, 29] is another possible candidate of the present mechanism, if one applies a parallel magnetic field to it. The antiferromagnetic long-range order with weak ferromagnetism due to the canted spin structure has been observed in this compound at zero field [29].

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[15] According to the theory in ref. [14], the electron states are nondegenerate almost everywhere in the momentum space because of strong spin-orbit coupling, and Cooper pairing occurs between electrons with $(\boldsymbol{k}, n)$ and $(-\boldsymbol{k}, n)$, which have the same energy $\epsilon_n(\boldsymbol{k}) = \epsilon_n(-\boldsymbol{k})$, where n denotes the band index. Since the states with $(\boldsymbol{k}, n)$ and $(-\boldsymbol{k}, n)$ are connected by the time reverse transformation, they cannot be equal spin states. If the state with $(\boldsymbol{k}, n)$ is up-spin rich, the state with $(-\boldsymbol{k}, n)$ must be down-spin rich. Thus, the pair of the electrons with $(\boldsymbol{k}, n)$ and $(-\boldsymbol{k}, n)$ experiences the Pauli paramagnetic pair-breaking effect. In ref. [30], it is shown that the paramagnetic suppression of superconductivity is nonzero in all directions in general, and the Knight shift has an unusual anisotropy of the temperature dependence.
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[17] We define the coupling constant J_{ij} as one including the corrections from the conduction electrons. In actuality, the influence of the conduction electron may not be included completely only through such corrections, because various spin structures other than the canted structure can occur [31]. However, we neglect such possibilities phenomenologically, and consider only the cases in which the magnetic subsystem is effectively described by eq. (1).

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[23] In the present scale, the Pauli limit $H_P = \Delta_0/\sqrt{2}\mu_B$ at $T = 0$ corresponds to $a_m H_P = h_P/2r_m\pi T_c^{(0)} = g_e/4\sqrt{2}e^{\gamma}r_m$, where $\gamma = 0.57721\cdots$. Therefore, we obtain $a_m H_P \approx 0.2/r_m$, if $g_e \approx 2$.

[24] It was observed that $H_{cent} \gtrsim 14\,\mathrm{T}$ and $H_{AF} \approx 5\,\mathrm{T}$ [7], which give $J_K \gg J'_{AF}$. Furthermore, since the FISC was observed for $H \gtrsim 14\,\mathrm{T}$, $H_{c20}(0) \gg 14\,\mathrm{T}$ must be satisfied, while $H_P \gtrsim 1.86 \times 0.6 \sim 1.1\,\mathrm{T}$, since $T_c^{(0)} \gtrsim 0.6\,\mathrm{K}$. Hence, we obtain $r_m \sim 0.2H_{c20}/H_P \gg 3 \gg 1$.

[25] If J_K's are on the same order in the compounds $CePb_3$ and $CePt_3Si$, it is expected that $H_{cent} \gtrsim 14\,\mathrm{T}$ in the compound $CePt_3Si$. Furthermore, comparing the values of $T_c^{(0)}$ and $T_{AF}^{(0)}$ of the two compounds, it seems plausible that $H_{c20}(0)$ should be much larger than 14 T, and that $H_{AF} \sim 10\,\mathrm{T}$. In this case, the condition $H_{c20}(0) > H_{cent} > H_{AF}$ is satisfied.

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